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DEFLAGRATIONO-DETONATION TRANSITION IN HMX-BASED PROPELLANTS.(U)

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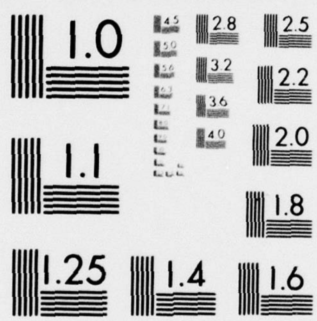

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The long-range objective of the research is to develop the capability of assessing the deflagration-to-detonation transition (DDT) hazard in HMX-based propellants. The approach is based on the concept that a basic understanding of the physical and chemical processes involved in DDT is necessary to achieve this objective. Hence, theoretical and experimental studies were undertaken to elucidate mechanisms of DDT, to establish conditions for its occurrence, and to formulate a satisfactory model for its quantitative description. The theoretical studies were concerned with the thermo-hydrodynamic treatment of DDT. The central

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DEFLAGRATION-TO-DETONATION TRANSITION IN HMX-BASED PROPELLANTS

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ABSTRACT

The long-range objective of the research is to develop the capability of assessing the deflagration-to-detonation transition (DDT) hazard in HMX-based propellants. The approach is based on the concept that a basic understanding of the physical and chemical processes involved in DDT is necessary to achieve this objective. Accordingly, theoretical and experimental studies were undertaken to elucidate mechanisms of DDT, to establish conditions for its occurrence, and to formulate a satisfactory model for its quantitative description.

The theoretical studies were concerned with the thermo-hydrodynamic treatment of DDT. The central problem is to model the propagation of the unsteay flame and the flow it produces ahead of the flame front. The flame was treated as a reactive discontinuity and flow conditions for propagation of steady and unsteady flames were established. Equations for the initial flow produced by the onset of deflagration in a closed tube were derived to demonstrate that an accelerating flame produces a compression wave ahead of the flame front. The initial attempt to construct an explicit solution for an accelerating flame in a closed tube, based on the group of transformations admitted by the flow equations, was unsuccessful.

The experimental studies were delayed by problems of finding a source of propellant samples suitable for Lagrange gage experiments. A source of HMX-based propellant was found at Edwards Air Force Base, and experimental procedures were changed to eliminate the machining and grooving operations usually employed in constructing assemblies for Lagrange gage experiments. A target assembly was designed to allow incorporation of the gages and ionization pins into the propellant charges during the casting process.

I INTRODUCTION

Research on the deflagration-to-detonation transition (DDT) is important to the Air Force because many of the energetic propellants required for present and future long-range delivery systems are explosively filled compositions that are capable of undergoing DDT. HMX-based propellant, for example, usually burns reliably in rocket motors but has detonated and destroyed the motor on several occasions. Possible steps leading to detonation in the rocket motor are fracture of propellant ahead of the flame and the subsequent formation of shock waves produced by the increased burning rate of fractured propellant. But even in this case, the mecahnism of DDT and the conditions for its initiation are not adequately understood.

The long-range objective of the present research program is to develop a computational capability for assessing the DDT hazard in explosively filled propellants. The program is based on the concept that a basic understanding of the physical and chemical processes involved in DDT is necessary to achieve this objective. Combined theoretical and experimental studies to determine the pressure fields behind and ahead of the flame and to establish mechanisms of DDT are required to develop such understanding.

II THEORETICAL STUDIES

The first task in the theoretical study is to develop a basic physical understanding of the thermo-hydrodynamic processes governing the flame and its propagation. Specific steps undertaken to attain such an understanding are as follows:

(1) Choose a model for the flame

(2) Derive and physically interpret the equations governing flame propagation

(3) Construct solutions for the unsteady flow produced by a flame propagating in a closed tube.

It is convenient first to choose the simplest model and treat the flame as a reactive discontinuity. Implicit in this choice are the assumptions that the burning reaction is fast and that the hydrodynamic flow produced by an unsteady flame is not appreciably influenced by the detailed structure of the flame. In this case, states connected by the burning process are governed by the Rankine-Hugoniot jump conditions expressing the conservation of mass, momentum, and energy across the reactive discontinuity.

Let v denote specific volume; F, flame velocity; u, particle velocity; p, pressure; c, sound speed; and h, specific enthalpy; and let the subscripts i and f denote the initial and final states, respectively connected by the flame. The states across the discontinuity are related by the Rankine-Hugoniot jump conditions

$$v_f(F-u_i) = v_i(F-u_f) \tag{1}$$

$$(u_f - u_i)^2 = (p_f - p_i)(v_i - v_f) \tag{2}$$

$$2(h_f - h_i) = (p_f - p_i)(v_f + v_i) \tag{3}$$

Equation (1) expresses the conservation of mass, Eq. (2) expresses the conservation of mass and momentum, and Eq. (3) expresses the conservation of mass, momentum, and energy. The locus of states attainable from a given initial state (p_i, v_i, h_i) lie on a curve in the (p,v) plane obtained by eliminating h between Eq. (3) and the $h = h(p,v)$ equation of state of the burnt products. This curve, called the Hugoniot curve centered on (p_i, v_i), has two branches when the discontinuity is reactive.

When the reaction is exothermic, the branch where $p_f > p_i$ and $v_f \leq v_i$ is called the detonation branch, and the branch where $p_f \leq p_i$ and $v_f > v_i$ is called the deflagration branch. With the present model of the flame, burnt states are represented by points on the deflagration branch of the Hugoniot curve. Moreover, the maximum flame speed is defined by the Chapman-Jouguet (CJ) point where $F = u_f + c_f$, the flow is sonic, and the Rayleigh line through the initial state (p_i,v_i),

$$p - p_i = \left(\frac{F-u_i}{v_i}\right)^2 (v_i - v) \tag{4}$$

is tangent to the Hugoniot curve.

Equations for deflagration CJ parameters will be derived and then the differential equation governing the propagation of a reactive discontinuity will be formulated. Equation of state information must be specified to compute CJ parameters and states on the deflagration Hugoniot curve. Let the subscript o denote the standard state and let the superscripts x and p denote propellant and propellant products, respectively. The initial enthalpy of the propellant and the $h = h^p(p,v)$ equation of state of the products are written as

$$h_i^x = \Delta H_o^x + h_i^x (p_i,v_i) \tag{5}$$

and

$$h^p = H_o^p + \frac{kpv}{k-1} \tag{6}$$

where $H_o^p = \Delta H_o^p - k\, p_o v_o/(k-1)$, ΔH_o denotes the heat of formation, and k denotes the polytropic index of propellant products. The combination of Eqs. (3) and (6) gives the equation for the Hugoniot curve centered on $(p_i v_i)$ as

$$p_f(\mu v_f - v_i) + p_i(v_i + v_f) = 2\ q \tag{7}$$

where $\mu = (k + 1)/\ (k - 1)$ and $q = [(\Delta H_o^x - \Delta H_o^p) + h^x(p_i, v_i) + k\ p_o v_o/k-1]$.

At this stage it is convenient to neglect the $kp_o v_o/(k-1)$ term in q and introduce the nondimensional variables $p_f/p_i = P$, $v_f/v_i = V$, $q/p_i v_i = Q$ and $U^2 = (u_f - u_i)^2/p_i v_i$. Equations (2) and (7) can then be written as

$$(P-1)\ (1-V) = U^2 \tag{8}$$

and

$$P(\mu V-1) + (V+1) = 2Q \tag{9}$$

Using Eq. (4) and the definition of sound speed $c^2 = kpv$, the CJ condition $F-u_f = c_f$ can be written as

$$\frac{P-1}{1-V} = k\ \frac{P}{V} \tag{10}$$

The CJ parameters are obtained by solving Eqs. (8) - (10) and Eq. (1). The combination of Eqs. (8) - (10) leads to the following equations relating U and V to Q.

$$\mu U^2 + (\mu + 1) - 2Q = 0 \tag{11}$$

$$(1-V)^2 + 2B(1-V) - (\frac{\mu - 1}{\mu})\ B = 0 \tag{12}$$

where $B = (2Q/(\mu+1) - 1)$. The roots of Eqs. (11) and (12) give the particle velocities and volumes at the CJ points on the detonation and deflagration branches of the Hugoniot curve centered at $(p_i v_i)$. Solving Eq. (11) and converting back to dimensional variables gives the equation

$$u_f = u_i \pm \frac{1}{k+1}\ [2\{(k^2-1)q - k(k+1)p_i v_i)\}]^{1/2} \tag{13}$$

where the positive sign gives the particle velocity at the CJ point on the detonation branch of the Hugoniot and the negative sign gives the particle velocity at the CJ point on the deflagration branch.

Whereas liberation of chemical energy in a detonation produces mass motion directed toward the reaction front, the liberation of chemical energy in a deflagration produces mass motion directed away from the reaction front. Expanding the square root term in the equation for the roots of Eq. (12) gives the following equations for the CJ volumes on the detonation (14) and deflagration (15) branches as

$$\frac{v_f}{v_i} = \frac{k}{k+1} \tag{14}$$

and

$$\frac{v_f}{v_i} = \frac{2(k-1)Q}{k} - \frac{k}{k+1} \tag{15}$$

Rearranging Eq. (1) to give the equation

$$F - u_i = \frac{(u_f - u_i)}{(V-1)} \tag{16}$$

and eliminating $(u_f - u_i)$ by using Eq. (13) with $A^2 = 2((k^2-1)q - k(k+1)p_i v_i)$ gives the equation for the maximum deflagration velocity as

$$F - u_i = \frac{A}{(1 + A^2/k p_i v_i)} \tag{17}$$

The equation for the CJ deflagration pressure then follows from Eq. (9) as

$$P - 1 = -\frac{A^2/p_i v_i}{(k+1)\ (1 + A^2/k p_i v_i)} \tag{18}$$

when $A^2/kp_i v_i >> 1$, Eq. (17) gives the maximum deflagration velocity as

$$F - u_i = k\,\frac{p_i v_i}{A} \tag{19}$$

and the corresponding equation for the change in pressure across the discontinuity follows from Eq. (19) as

$$p_i - p_f = \frac{kp_i}{k+1} \tag{20}$$

Equations (13), (15), and (18) show that the reaction in a CJ deflagration wave produces a decrease in particle velocity and pressure but an increase in volume. Other states on the deflagration Hugoniot curve must be considered, however, because the CJ deflagration represents a hypothetical idealized case. These states lie on the weak deflagration branch where $p_{CJ} < p_f \leq p_i$, and where, according to Jouguet's rules, the flow of reaction products is subsonic with respect to the reaction front. Jouguet's rules for the flow in front of a weak deflagration give an insight into the problem of modeling unsteady deflagration and DDT. The flow ahead of the wave is influenced by the wave itself because the flow ahead of the wave is subsonic with respect to the wave front. Consequently, an accelerating flame produces an unsteady compression wave ahead of itself and the initial pressure for the burning process increases as the flow develops. The burnt states are represented by points on the family of deflagration Hugoniot curves centered on an adiabatic compression curve of unburnt material. Moreover, the continued acceleration of the flame will result in shock formation and initiation of detonation in material ahead of the flame front.

The differential equation governing the propagation of a reactive discontinuity will now be derived to obtain a more quantitative description of unsteady deflagration waves. Let t and x denote time and Eulerian distance, respectively, and $p_i - p_f = \Delta p$ and $u_i - u_f = \Delta u$. When the deflagration propagates as a discontinuity, the time variations of p_i and p_f satisfy the equations

$$\frac{Dp_i}{Dt} = \frac{\partial p}{\partial t_i} + F\,\frac{\partial p}{\partial x_i} \tag{21}$$

$$\frac{Dp_f}{Dt} = \frac{\partial p}{\partial t_f} + F\,\frac{\partial p}{\partial x_f} \tag{22}$$

where the subscripts i and f denote that the partial derivatives are evaluated at the top and bottom of the discontinuity. Subtracting Eqs. (21) and (22) after the partial time derivatives have been eliminated using the identity

$$\frac{dp}{dt} = \frac{\partial p}{\partial t} + u\,\frac{\partial p}{\partial x} \tag{23}$$

leads to the equation

$$\frac{D\Delta p}{Dt} = \frac{dp}{dt_i} - \frac{dp}{dt_f} + (F-u_i)\,\frac{\partial p}{\partial x_i} - (F-u_f)\frac{\partial p}{\partial x_f} \tag{24}$$

A similar procedure leads to the corresponding equation for u

$$\frac{D\Delta u}{Dt} = \frac{du}{dt_i} - \frac{du}{dt_f} + (F-u_i)\frac{\partial u}{\partial x_i} - (F-u_f)\frac{\partial u}{\partial x_f} \tag{25}$$

The equation governing the discontinuity is obtained by combining Eqs. (24) and (25) with the equations of motion, which are written as

$$\rho\frac{du}{dt} = -\,\frac{\partial p}{\partial x} \tag{26}$$

$$\frac{dp}{dt} = c^2\,\frac{d\rho}{dt} = -\,c^2\rho\,\frac{\partial u}{\partial x} \tag{27}$$

Multiplying Eq. (25) by $m = \rho_i(F-u_i) = \rho_f(F-u_f)$ and adding the resulting equation to Eq. (24) leads to the equation

$$\frac{D\Delta p}{Dt} + m\,\frac{D\Delta u}{Dt} = -\,\rho_i(c_i^2 - (F-u_i)^2)\,\frac{\partial u}{\partial x_i} + \rho_f(c_f^2 - (F-u_f)^2)\,\frac{\partial u}{\partial x_f} \tag{28}$$

Equation (28) gives hydrodynamic conditions associated with deflagration waves. Consider first a deflagration wave propagating at constant velocity when $D(\Delta p)/\Delta t = D(\Delta u)/Dt = 0$ and both terms on the right-hand side of Eq. (28) are zero. Necessary conditons for this case are either $\partial u/\partial x_i = \partial u/\partial x_f = 0$, or $\partial u/\partial x_i = 0$ and $c_f = F - u_f$, because unreacted material satisfies the condition $c_i > F - u_i$. Solutions involving a pre-compression shock followed by a deflagration wave can be constructed to satisfy both of these conditions. Rejection of the CJ condition on physical grounds, however, gives $\partial u/\partial x_i = \partial u/\partial x_f = 0$ as the necessary hydrodynamic condition for steady deflagration waves. Of particular interest is the flow when the particle velocity of burnt material is zero. In this case the strengths of the shock and deflagration waves are such that the increase in particle velocity produced by the shock is exactly offset by the decrease in particle velocity produced by the deflagration.

Now consider the case of an accelerating deflagration $D(\Delta p)/Dt > 0$, $D(\Delta u)/Dt > 0$ supporting a compression wave. Because $c_i > F-u_i$ and $\partial u/\partial x_i < 0$ in the compression wave, and $c_f > F-u_f$ in the products, the possibility arises that Eq. (28) can be satisfied when $\partial u/\partial x_f$ is positive, zero, and negative.

The flow produced by the propagation of a deflagration wave from a rigid wall is of particular interest because our experimental study of DDT is designed to model this rear-boundary condition. Equations for the initial flow will be derived for this case to show that a compression wave must be formed as the deflagration propagates from wall. Let the superscript o denote the initial condition at time t = 0. Then initially the rear-boundary conditon is expressed by the equations $u_f^o = (du_f/dt)^o = 0$. We also assume that the unburnt material is initially at rest, $u_i^o = 0$, and that the initial velocity of the flame is zero, $F^o = 0$. It follows from the jump conditions, Eqs. (1), (2), and (3), the $p_f^o = p_i^o$, and $h_f^o = h_i^o$, but that $\rho_f^o \ll \rho_i^o$. The equation relating the increase in volume to the energy liberated in the reaction can be readily derived by combining the constant enthalpy condition with Eqs. (5) and (6). The initial condition for propagation of the flame along the tube is $(DF/Dt)^o > 0$. Other initial conditions follow from the equations $m = \rho_i(F-u_i) = \rho_f(F-u_f)$, and $p_i - p_f = m(u_i - u_f)$, the identities for the pressure and particle velocity derivatives, and the flow equations, Eqs. (26) and (27). Differentiating m and imposing the initial and rear-boundary conditons leads to the equations

$$\frac{Dm^o}{Dt} = \rho_i{}^o \left(\frac{DF}{Dt} - \frac{Du_i}{Dt}\right)^o = \rho_f{}^o \; \frac{DF^o}{Dt} \qquad (29)$$

and rearranging Eq. (29) gives the equation

$$(\rho_i - \rho_f)^o \frac{DF^o}{Dt} = \rho_i^o \frac{Du_i^o}{Dt} \tag{30}$$

Because $\rho_i^o > \rho_f^o$, and $(Du_i/Dt)^o = (du_i/dt)^o$, it follows from Eq. (30) that $(Du_i/Dt)^o = (du_i/dt)^o > 0$ and that the particle velocity in front of the flame starts to increase as the flame starts to propagate along the tube. That is, the deflagration process produces a compression wave ahead of the flame front. Differentiating the momentum equation and imposing the initial and rear-boundary conditons leads to the equations

$$\frac{Dp_i^o}{Dt} = \frac{Dp_f^o}{Dt} = \frac{dp_i^o}{dt} = \frac{dp_f^o}{dt} \tag{31}$$

which shows that the initial time rate of change of pressure is the same in burnt and unburnt materials. The assumption that material in front of the flame is compressed along an isentrope is sufficient to determine other initial conditions. Combination of the isentropic condition $dp_i = (\rho c)_i \, du_i$ with Eqs. (30) and (31) gives the equation

$$\frac{dp_i^o}{dt} = \frac{dp_f^o}{dt} = c_i^o(\rho_i - \rho_f)^o \frac{DF^o}{Dt} \tag{32}$$

which shows that the pressure starts to increase behind and in front of the flame as the flame starts to propagate along the tube. The corresponding equations for the particle velocity gradients obtained by combining Eq. (32) with the energy equation $dp/dt = -\rho c^2 \, \partial u/\partial x$ show that the initial values of the particle velocity gradients behind and in front of the deflagration are negative.

Initial attempts to use group theoretical methods to construct solutions for the development of the flow produced by the propagation of a deflagration wave in a closed tube have been unsuccessful.

EXPERIMENTAL STUDIES

The experimental studies have been delayed by problems of finding a source of propellant for the Lagrange gage experiments. Protracted negotiations to obtain HMX-based propellant from Lawrence Livermore Laboratory were finally unsuccessful for the following reasons:

- Propellant was in short supply
- The machining costs and times to meet tolerances in the dimensions of propellant samples needed to construct the DDT gage assemblies were prohibitive.

We subsequently found a source of HMX propellant at Edwards Air Force Base (EAFB), and personnel there agreed to provide the propellant required for this program. After discussions with EAFB personnel, we concluded that the usual method of constructing targets for Lagrange gage experiments from machined pieces was not practicable for HMX-based propellants. Hence, a target assembly based on casting rather than machining the propellant was designed to eliminate the complex machining and grooving operations conventionally used in constructing targets for Lagrange gage experiments. Target assemblies consisting of acrylic tubes containing the stress gages and ionization pins are being constructed and will be sent to EAFB to be filled with propellant as soon as they are completed. Thick-wall steel confinement tubes to contain these target assemblies are being constructed at SRI. We expect to assemble and perform the initial DDT experiments shortly after we receive the cast charges from EAFB.

FUTURE WORK

Theoretical and experimental studies will be continued during the second year of the contract to model and establish conditions for the onset and occurrence of DDT in HMX-based propellants. Solutions for the unsteady flow produced by a deflagration will be sought to provide a basis for interpreting the results of the DDT experiments. The pressure histories recorded in the DDT experiments will be incorporated into the treatment of the unsteady flame to generate a satisfactory model for DDT.